

Deer Park, TX - Spatial Analysis of Volatile Organic Compounds from a Community-Based Air Toxics Monitoring Network

Phase IV of PASSIVE AMBIENT AIR TOXICS MONITORING IN THE HOUSTON-GALVESTON AREA

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The project has five phases as listed below. This report contains Phase IV. The final reports can be found at US/EPA Website under:

<http://www.epa.gov/ttn/amtic/files/ambient/passive>.

Phase I. Determination of Optimum Sampling Conditions

Phase II. Year-long Temporal Passive Monitoring at High and Low TRI Emissions; Aldine and Clinton areas

Phase III. Community-based Spatial Monitoring; Aldine, Clinton, and Deer Park

Phase III Supplementary Monitoring

Phase IV. Spatial Analysis of Passive VOCs from the Community-Based Network in Deer Park near Houston Ship Channel, TX

Phase V. Statistical Analysis of Passive VOCs in Aldine and Clinton near Houston, TX

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ABSTRACT

In the summer of 2003, ambient air concentrations of volatile organic compounds (VOCs) were measured at twelve sites within a 3-km radius in Deer Park, Texas near Houston. The purpose of the study was to assess local spatial influence of traffic and other urban sources and was part of a larger investigation of VOC spatial and temporal heterogeneity influences in selected areas of Houston. Seventy two-hour samples were collected using passive organic vapor monitors. Most measurements of 13 VOC species were greater than the method detection limits. Samplers were located at 10 residential sites, a regulatory air monitoring station, and a site located at the centroid of the census tract in which the regulatory station was located. For residential sites, sampler placement locations (e. g., covered porch vs. house eaves) had no effect on concentration with the exception of methyl tertiary-butyl ether (MTBE). Relatively high correlations (Pearson $r > 0.8$) were found between toluene, ethylbenzene, and o,m,p-xylenes suggesting petroleum-related influence. Chloroform was not correlated with these species or benzene (Pearson $r < 0.35$) suggesting a different source influence, possibly from process-related activities. As shown in other spatial studies, wind direction relative to source location had an effect on VOC concentrations.

Keywords: air pollution, passive samplers, refineries, traffic, spatial analysis, volatile organic compounds (VOC), wind direction

INTRODUCTION

Spatial analysis studies of ambient air pollutants have been conducted to assess traffic influence and its possible impact on respiratory health (van Vliet et al., 1997; Roorda-Knappe et al., 1998; Brauer et al., 2003). Similar research has been enhanced by the incorporation of ancillary variables from geographic and demographic sources to assess neighborhood differences of air pollution; this approach has been term as land-use regression modeling (Jerrett et al., 2005; Gilbert et al., 2005).

Conventional, regulatory-based air monitoring is generally expensive and, thus, conducted at one or a few select locations in a city. This provides limited information on intra-urban variability for air pollution. Research-oriented urban network monitoring has progressed with the increased use of passive diffusion sampling and advances in sampler portability.

For a better understanding of ambient air toxics impacts in the Houston metropolitan area, the Region 6 office of the U.S. Environmental Protection Agency (EPA) and the University of Texas-Houston School of Public Health conducted extensive spatial and temporal ambient VOC monitoring using passive sampling devices for VOCs known as organic vapor monitors (OVMs) in select census tracts (Stock et al, 2005, Stock et al., 2005). A community-based air sampling approach at 31 homes, 3 state monitoring sites, and 3 census tract centroids in three distinct locations in Houston were conducted as

part of that assessment. Spatial differences were apparent between neighborhood sites and their corresponding central locations (Stock et al., 2005). The analyses presented here shall focus on measurements from the Deer Park area.

METHODS

Deer Park, Texas was selected since it is in the vicinity of the Houston Ship Channel and has a relatively high density of chemical industry point and mobile sources of air toxics. The Deer Park residential sites were also considered to be less impacted by immediate activities that could adversely affect spatial assessments of overall ambient influences (Stock, personal communication). The study design, sampling methodology, and chemical analyses are discussed elsewhere (Stock et al., 2004). Aspects of the study relevant to the analyses reported here are briefly mentioned. Passive samplers for VOCs in $\mu\text{g}/\text{m}^3$ (Model 3500 OVM; 3M Company, St. Paul, MN) were located at 10 residential sites, a regulatory continuous air monitoring station operated by the State of Texas, and a site centrally located in the census tract in which the monitoring station was located. The OVM passive sampler has been validated in other studies under controlled, ambient and exposure conditions in this urban area and elsewhere (Chung et al., 1999; Mukerjee et al., 2004; Pratt et al., 2005; Weisel et al., 2005) and has been used extensively elsewhere (Cohen et al., 1990; Ware et al., 1993; Sexton et al., 2004).

The OVM sampling locations were as follows: under eaves at four houses, in a porch area at five houses, above a garage door at the back of a carport at one house, a covered metal sampling stand on the roof of a trailer at the State of Texas site, and the back of a traffic sign at the central site. Five homes had collocated OVMs.

Monitoring occurred from May 20 to September 6, 2003; a total of six sampling periods were conducted, each period being a 72 h time integral. Recorded start and stop times indicated samples were placed and retrieved between 2:00 and 4:00 PM during each sampling event. Table 1 presents the VOC species analyzed from the OVMs and their below detection limit (BDL) counts. Species measured included petroleum-dominated compounds such as benzene, toluene, ethylbenzene, o- and m,p-xylenes (collectively referred to as BTEX species) and process-related compounds such as methyl tertiary-butyl ether (MTBE), chloroform, carbon tetrachloride, and BTEX species (Ware et al., 1993; Henry et al., 1997; Fujita et al., 2001). Petroleum dominated sources include transportation and refineries in the Houston Ship Channel area. MTBE is typically a gasoline additive (Cooney, 2005) but was also considered a process-related tracer for this study owing to a large MTBE production facility in Deer Park, according to the EPA Toxic Release Inventory for 2002.

The largest number of BDL counts was observed for toluene. It was confirmed that the BDL counts reported in Table 1 were reasonable (Stock, personal communication). The BDL values were set to half the detection limit. Values reported from collocated samplers were averaged for each sampling period.

Hourly meteorological data (wind speed, wind direction, temperature, and relative humidity) from the regulatory site were extracted from the EPA's Air Quality System (AQS). The hourly meteorological data were summarized on the same time frames as the passive monitoring sampling periods. This was done by assuming that all the OVMs were put in place at 2:00 PM during each of the six sampling periods and retrieved after the reported sampling duration. Resultant wind directions were averaged as vector quantities (Saucier, 1955; Mardia and Jupp, 2000).

Preliminary Analysis

Inspection of the data detected two anomalies. In one case, a sample had an anomalously low duration. After consultation with one of the investigators of the larger project, a correction factor was applied to the data from this sample (Stock, personal communication). The other suspicious element found in the reported chemical data set was that a relatively large discrepancy between collocated samplers was reported at one house during two sampling periods. The discrepancy existed for almost all the VOCs examined; no other similar difference existed at any other site or for this site during other sampling periods. Figures 1—7 show this discrepancy for BTEX compounds and chloroform and carbon tetrachloride for the house from which the suspicious collocated samples were taken. Review of site location information indicated that there was a potential influence on one of the collocated samplers due to its placement (Stock, personal communication). For this reason, the large chemical concentrations reported from this residence for these periods were deleted from the subsequent analyses.

RESULTS

Table 2 displays the correlations among the different VOC species; both Pearson and Spearman correlation coefficients are provided. Using the criterion of a Pearson coefficient of $r \geq 0.80$, each BTEX species was highly correlated with the other BTEX species, except benzene. Benzene was highly correlated only with toluene ($r=0.85$). Among the other chemicals, methylcyclopentane was highly correlated with MTBE ($r=0.87$), n-hexane ($r=0.88$), ethylbenzene ($r=0.80$), and m,p-xylene ($r=.83$). Ethylbenzene and n-hexane were also highly correlated ($r=0.80$), as were m,p-xylene and MTBE ($r=0.80$). The trimethylbenzenes were highly correlated with each other and each BTEX species except benzene (and toluene for 1,3,5-trimethylbenzene). The correlation results for BTEX species and these other compounds suggest a petroleum-related impact.

Generally, the same associations were observed with both the Pearson and Spearman correlation coefficients. However, the Spearman correlation coefficients suggested a stronger relationship between the BTEX species and MTBE and methylcyclopentane. Neither correlation measure suggested much relationship between n-pentane, carbon tetrachloride, or chloroform and any other pollutant examined. While further measurements would be necessary the low correlation of these species may suggest other sources such as process-related (e.g., industrial) influences. Chloroform and carbon tetrachloride are considered tracers-of-opportunity for process-related influences (Ware et

al., 1993); chloroform is also known to be emitted from waste water (Graedel, 1978).

Since different mounting placements were used at the residences, the eave locations were compared to the porch locations. The Wilcoxon rank sum test indicated no significant difference between these two mounting locations for any VOC; all p-values were greater than 10%. The Kruskal-Wallis test (Hollander and Wolfe, 1999) was conducted to check for overall differences among the eave, porch, carport, TCEQ, and central locations. Of the thirteen tests done, a statistically significant difference was found only for MTBE ($p=0.05$). Follow-up testing with the Wilcoxon rank sum test (Hollander and Wolfe, 1999) suggested the carport location to be significantly ($p<0.05$) higher than either the porch or State of Texas locations for MTBE.

Generally, the meteorological data showed little variability across sampling periods. All sampling periods were characterized by calm winds, high temperature, and high humidity, which is typical of the region. During the six periods of the study, average wind speeds ranged from 10.2 kph to 5.7 kph, average temperatures lay between 25.3 °C and 29.2 °C, and the average relative humidity levels were between 63% and 77%. The sole exception to this was for the resultant wind direction. For periods 1 and 6 the winds were predominantly blowing from the north, and for periods 2, 3, and 4 the winds were primarily blowing from the south. In period 5 the winds were virtually evenly divided between the southeast and northwest directions. Figures 8—13 plot the resultant hourly wind directions on the unit circle for each sampling period.

To examine whether the chemical concentrations differed with wind direction, the periods with northerly wind direction were compared to those with southerly wind direction using the Wilcoxon rank sum test. The results indicated that all the VOCs tested had higher concentrations when winds were from the north, with the following exceptions. Carbon tetrachloride had higher concentrations when winds were from the south, and there was no significant difference in chloroform concentration with wind direction. These findings are summarized in Table 3 for all VOCs and displayed for select VOCs in Figures 14—21. (The bar graphs in these figures are placed at the residential locations and the explicitly identified centroid and TCEQ sites.)

As displayed in the emissions insert of these figures, most of the emission sources for each pollutant lie to the north of the Deer Park area. These include both point sources, and for the vehicle-related pollutants, State Route 225. Except for carbon tetrachloride and chloroform, the fact that concentrations were higher when winds were from the north is consistent with the location of these emission sources relative to the Deer Park area. Prior to data analysis, it was suspected that the Sam Houston Parkway (Beltway Route 8) to the west of the Deer Park area would have an influence on the sites; however, wind directions during the sampling periods were such that no effect from that freeway was evident.

CONCLUSIONS

Passive sampling data for thirteen VOCs from the Deer Park area of Houston,

Texas were examined in this analysis. Relatively high correlations of BTEX species suggest a petroleum-related (traffic, refinery, etc.) influence. The BTEX species also had low correlations with chloroform and carbon tetrachloride.

Sampler placement with respect to porch vs. eave location had no effect on concentration. While immediate influences need to be considered for such network monitoring efforts, this suggests for the Deer Park data that differences in outdoor location relative to the residences themselves may not have been a major factor in terms of sampler variability. There was some indication that MTBE might be higher at the carport location as opposed to porch placement or the State of Texas site.

During the study, wind direction had a significant effect on concentration of the VOCs. Consistent with the location of the emission sources (including a freeway for the transportation-related species), most chemicals had higher concentrations when winds were from the north, though carbon tetrachloride concentrations were higher when winds were from the south. Previous respiratory health studies examining impact from traffic sources have suggested associations of health effects with gaseous pollutants such as nitrogen dioxide for subjects living downwind and within 100 m from freeways (van Vliet et al., 1997). Ultrafine particulate matter measurements conducted near freeways in Los Angeles have also suggested that ultrafine levels at immediate locations (30 m) downwind of freeways have different composition than locations 300 m downwind or upwind (Zhu et al., 2002). Although further studies are necessary, the VOC measurements here may be confirming the dominance of downwind influence relative to source location. Obviously, directional analysis to determine where pollutants may be coming from needs to be factored into spatial assessments from air monitoring network data.

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Chemical	BDL count (of 101 observations)
Benzene	0
Toluene	44
Ethylbenzene	0
m,p-xylene	0
o-xylene	1
Chloroform	4
carbon tetrachloride	0
n-pentane	11
MTBE	1
n-hexane	13
methylcyclopentane	5
1,3,5-trimethylbenzene	13
1,2,4-trimethylbenzene	0

Table 1. Below detection limit counts for the VOCs.

	V1	V2	V3	V4	V5	V6	V7	V8	V9	V10	V11	V12	V13
V1	1	.85	.75	.63	.67	.28	-.26	.46	.58	.70	.57	.53	.58
V2	.87	1	.92	.85	.88	.26	-.52	.39	.73	.78	.76	.78	.84
V3	.83	.92	1	.95	.94	.30	-.45	.43	.77	.80	.80	.84	.85
V4	.68	.79	.91	1	.97	.31	-.47	.40	.80	.79	.83	.83	.83
V5	.70	.83	.94	.94	1	.34	-.48	.30	.78	.74	.78	.84	.86
V6	.14	.15	.27	.33	.34	1	.02	.03	.17	.20	.08	.16	.14
V7	-.41	-.54	-.47	-.51	-.52	-.02	1	-.18	-.43	-.54	-.57	-.58	-.60
V8	.61	.46	.44	.44	.30	-.08	-.20	1	.38	.76	.59	.28	.23
V9	.70	.74	.81	.81	.84	.31	-.49	.34	1	.76	.87	.71	.71
V10	.85	.82	.81	.81	.77	.18	-.56	.70	.80	1	.88	.69	.69
V11	.78	.80	.79	.85	.78	.16	-.64	.60	.82	.91	1	.76	.75
V12	.60	.73	.82	.80	.86	.23	-.57	.22	.78	.65	.72	1	.96
V13	.69	.77	.84	.79	.87	.19	-.58	.24	.77	.68	.74	.96	1

Table 2. Correlation coefficients among the Deer Park VOCs. Pearson coefficients are in the upper triangular portion of the table and Spearman coefficients in the lower triangular section.

Key:

V1- benzene	V8 - n-pentane
V2 - toluene	V9 - MTBE
V3 - ethylbenzene	V10 - n-hexane
V4 - m&p-xylenes	V11 - methylcyclopentane
V5 - o-xylene	V12 - 1,3,5-trimethylbenzene
V6 - chloroform	V13 - 1,2,4-trimethylbenzene
V7 - carbon tetrachloride	

Wind Direction With Higher Concentrations	Chemical	P-value
North	benzene	<0.0001
	toluene	<0.0001
	ethylbenzene	<0.0001
	m&p-xylenes	0.0002
	o-xylene	<0.0001
	n-pentane	0.0021
	MTBE	<0.0001
	n-hexane	<0.0001
	methylcyclopentane	<0.0001
	1,3,5-trimethylbenzene	<0.0001
	1,2,4-trimethylbenzene	<0.0001
South	carbon tetrachloride	0.0011
No Difference	chloroform	0.2076

Table 3. Comparison of concentrations with respect to wind direction.

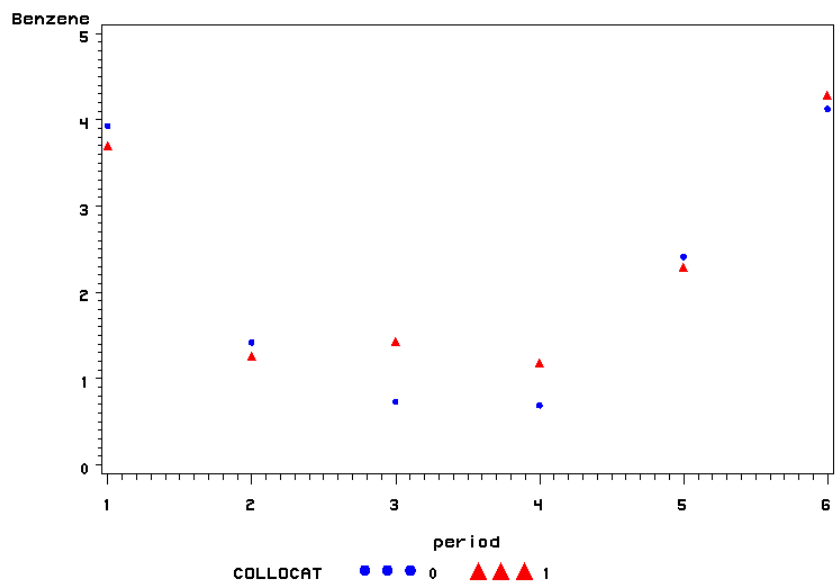


Figure 1. Benzene concentrations vs. sampling period.

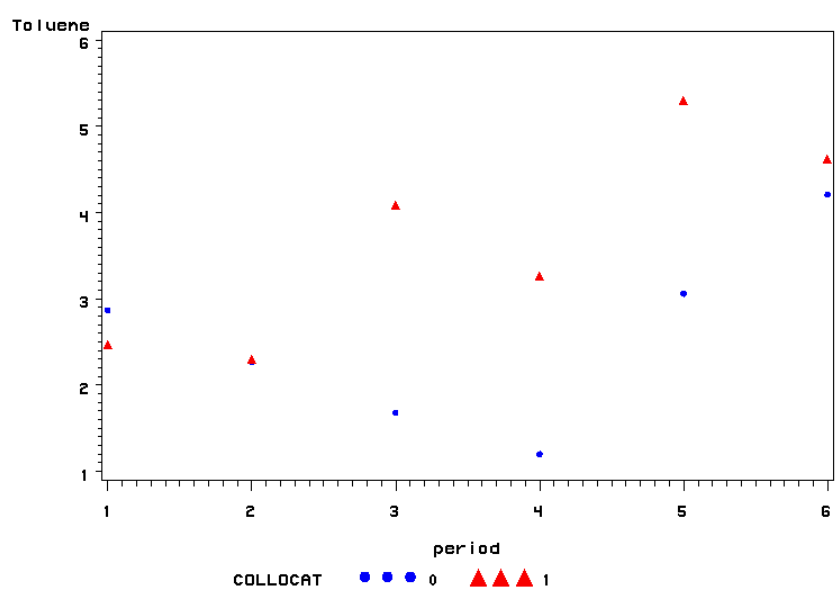


Figure 2. Toluene concentrations vs. sampling period.

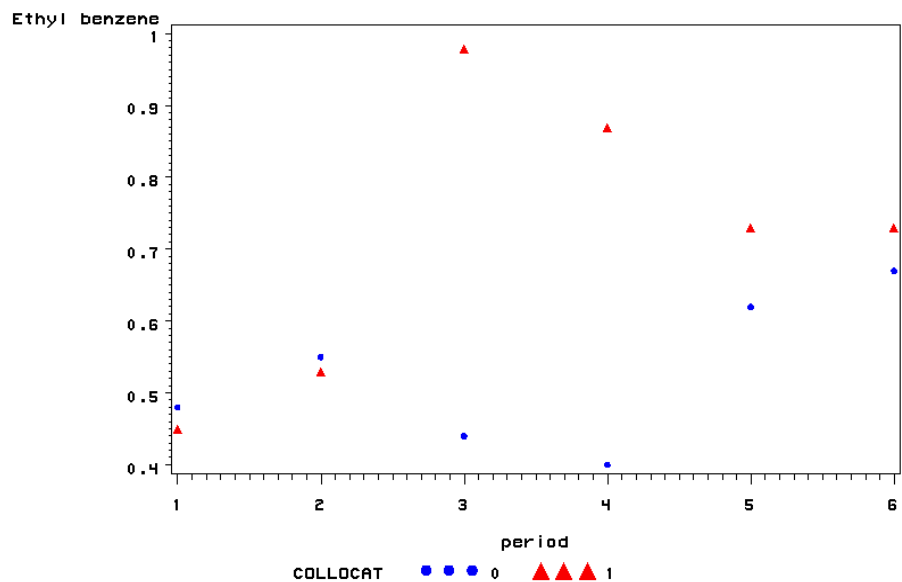


Figure 3. Ethylbenzene concentrations vs. sampling period.

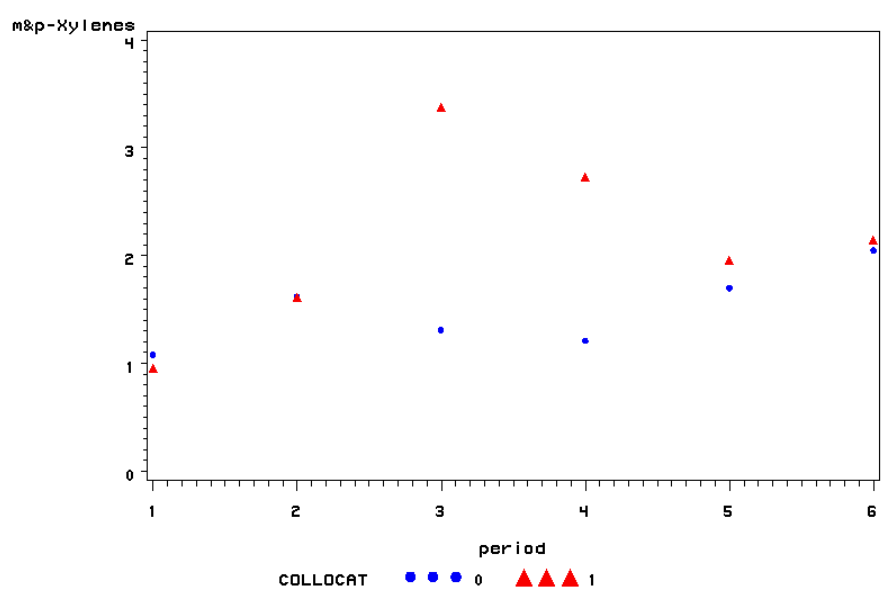


Figure 4. m&p-xylene concentrations vs. sampling period.

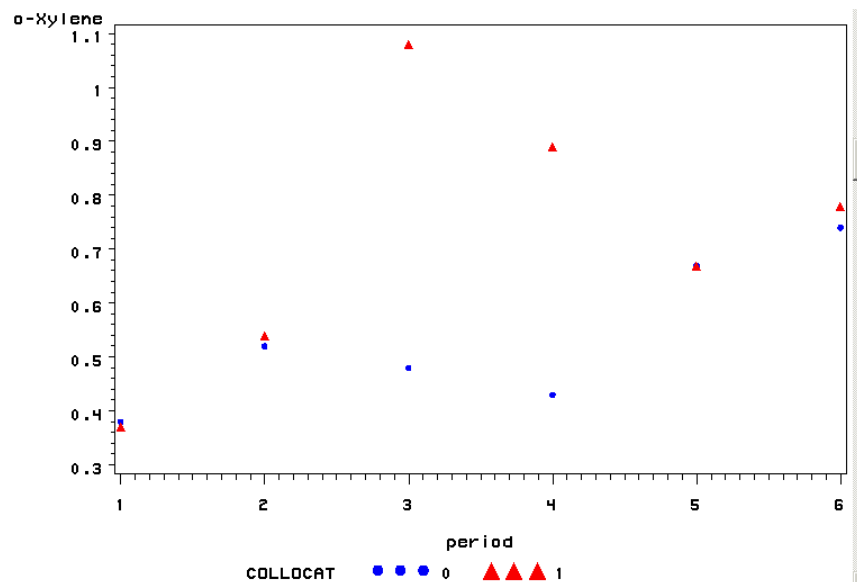


Figure 5. o-xylene concentrations vs. sampling period.

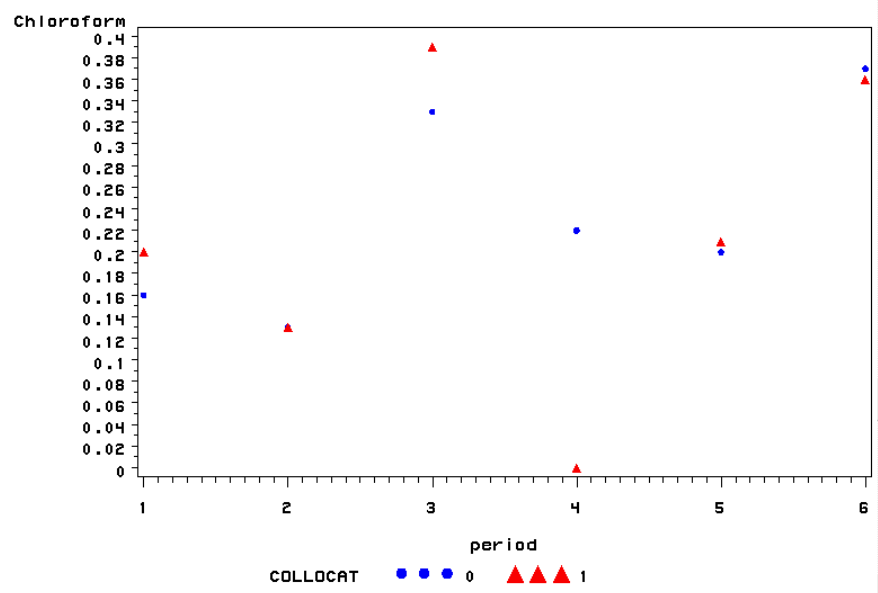


Figure 6. Chloroform concentrations vs. sampling period.

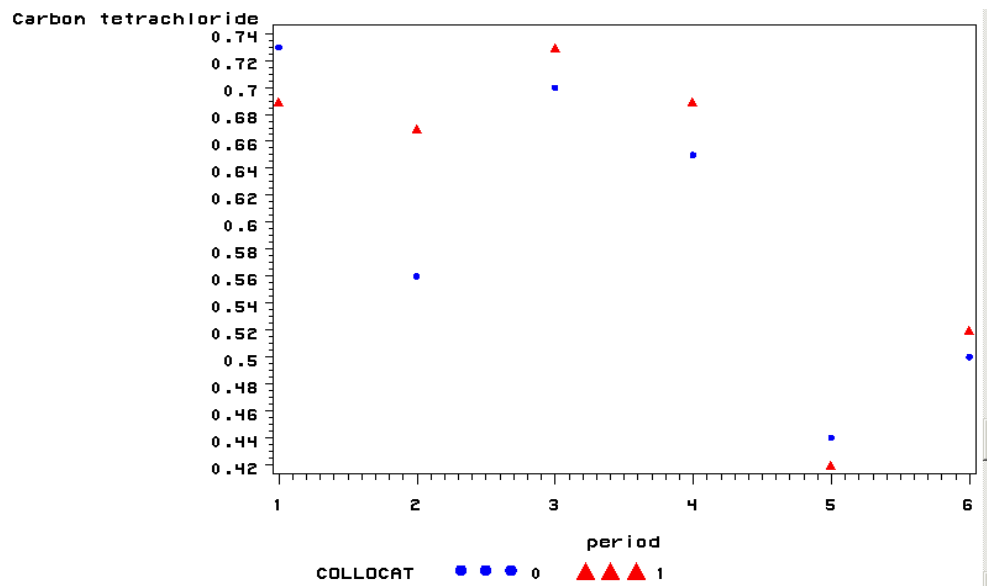


Figure 7. Carbon tetrachloride concentrations vs. sampling period.

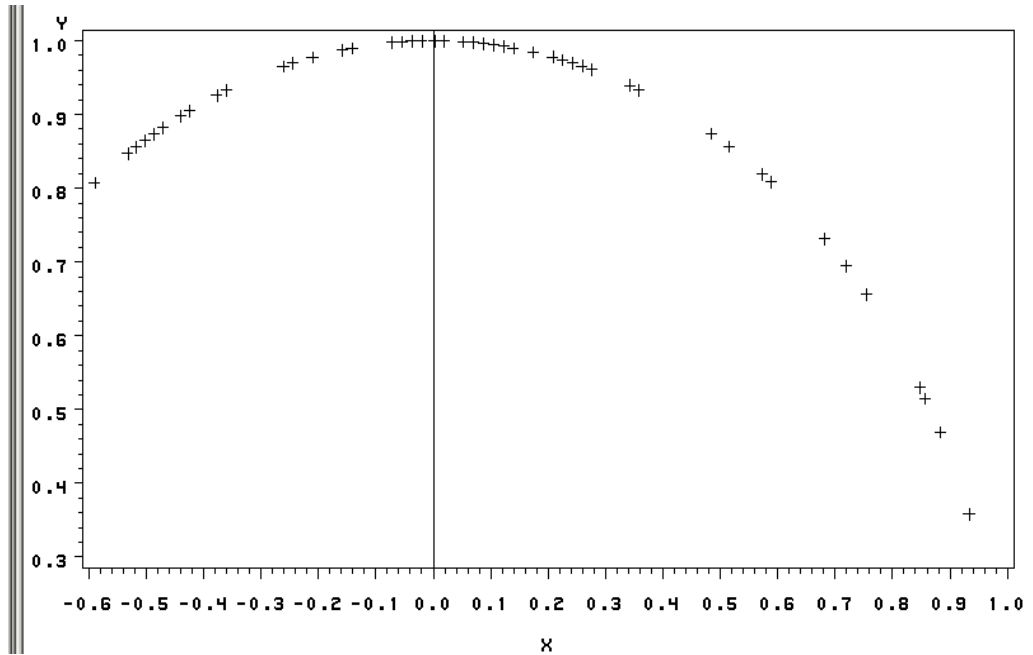


Figure 8. Resultant hourly wind directions for period 1. The (x,y) axes are interpreted as: (0,1) – due north; (1,0) – due east; (0,-1) – due south; (-1,0) – due west. (Data were only reported from the northeast and northwest quadrants).

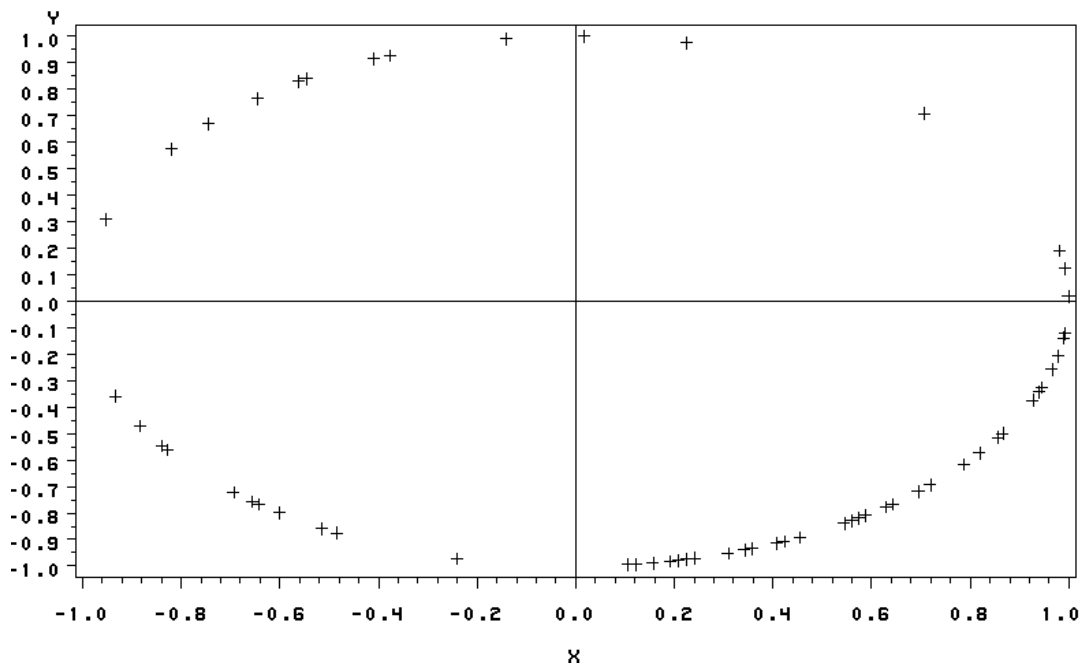


Figure 9. Resultant hourly wind directions for period 2. The (x,y) axes are interpreted as: (0,1) – due north; (1,0) – due east; (0,-1) – due south; (-1,0) – due west.

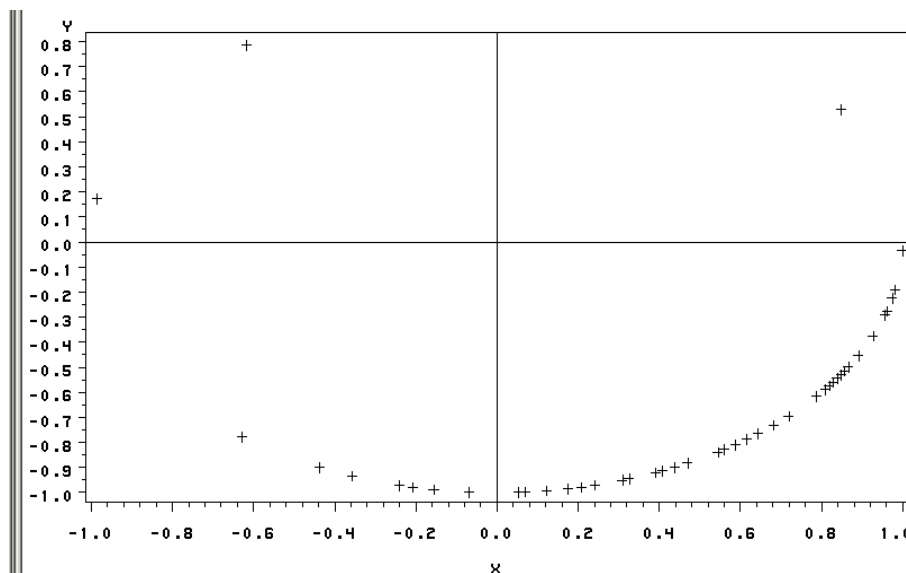


Figure 10. Resultant hourly wind directions for period 3. The (x,y) axes are interpreted as: (0,1) – due north; (1,0) – due east; (0,-1) – due south; (-1,0) – due west.

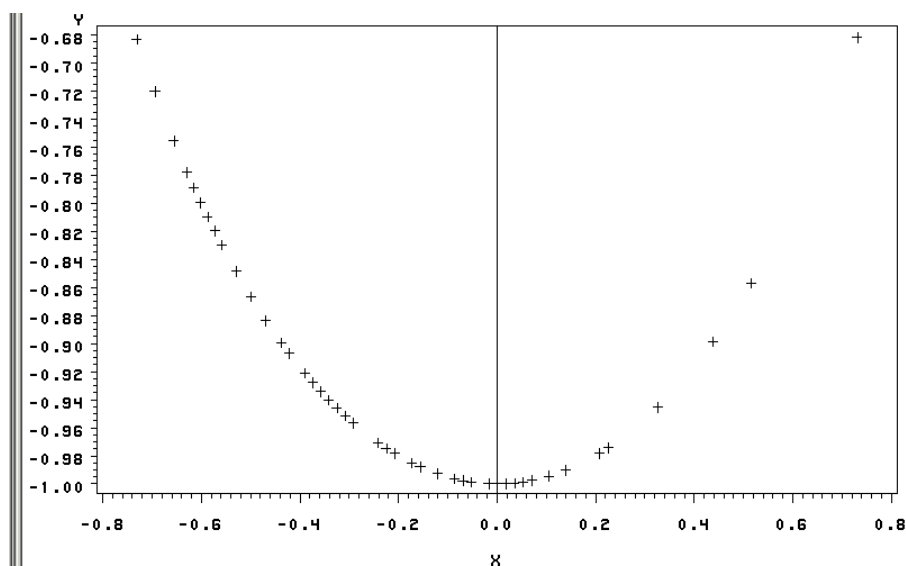


Figure 11. Resultant hourly wind directions for period 4. The (x,y) axes are interpreted as: (0,1) – due north; (1,0) – due east; (0,-1) – due south; (-1,0) – due west. (Data were only reported from the southeast and southwest quadrants).

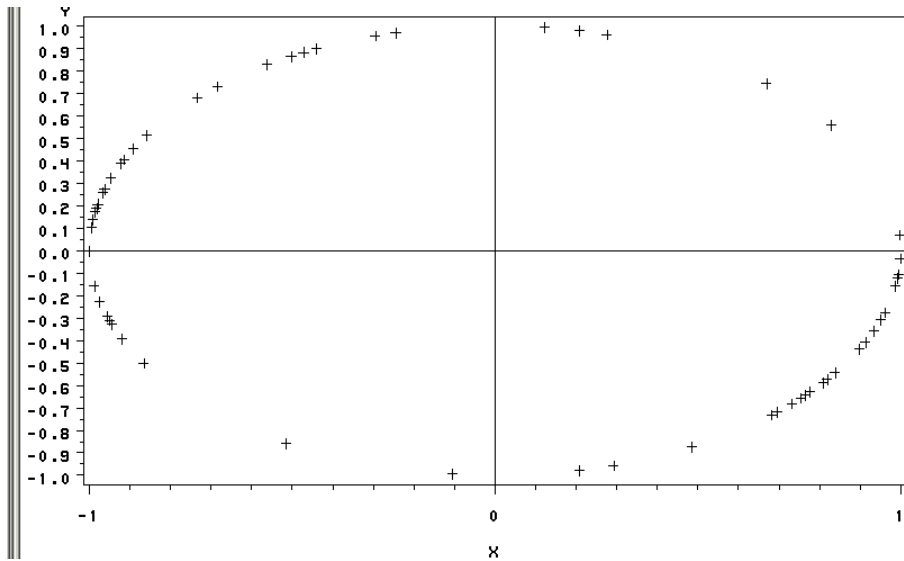


Figure 12. Resultant hourly wind directions for period 5. The (x,y) axes are interpreted as: (0,1) – due north; (1,0) – due east; (0,-1) – due south; (-1,0) – due west.

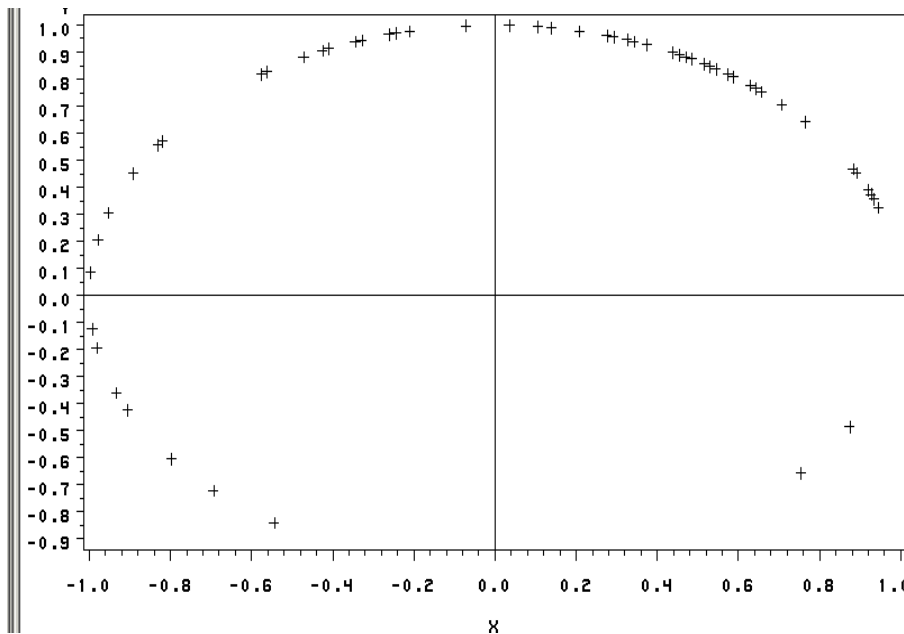


Figure 13. Resultant hourly wind directions for period 6. The (x,y) axes are interpreted as: (0,1) – due north; (1,0) – due east; (0,-1) – due south; (-1,0) – due west.

Benzene Concentrations in Deer Park

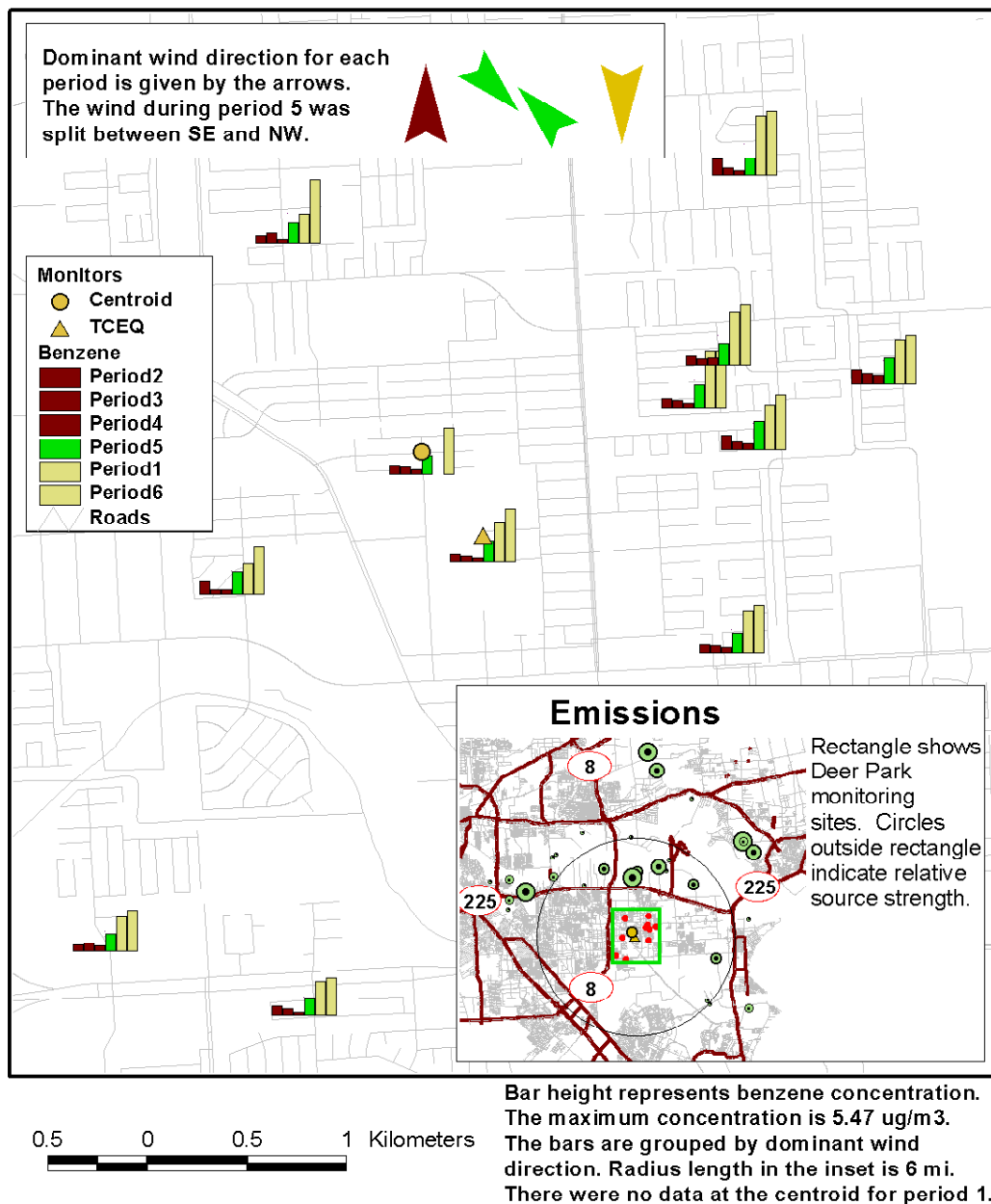


Figure 14. Benzene concentrations and emission sources in the Deer Park area of Houston, Texas.

Toluene Concentrations in Deer Park

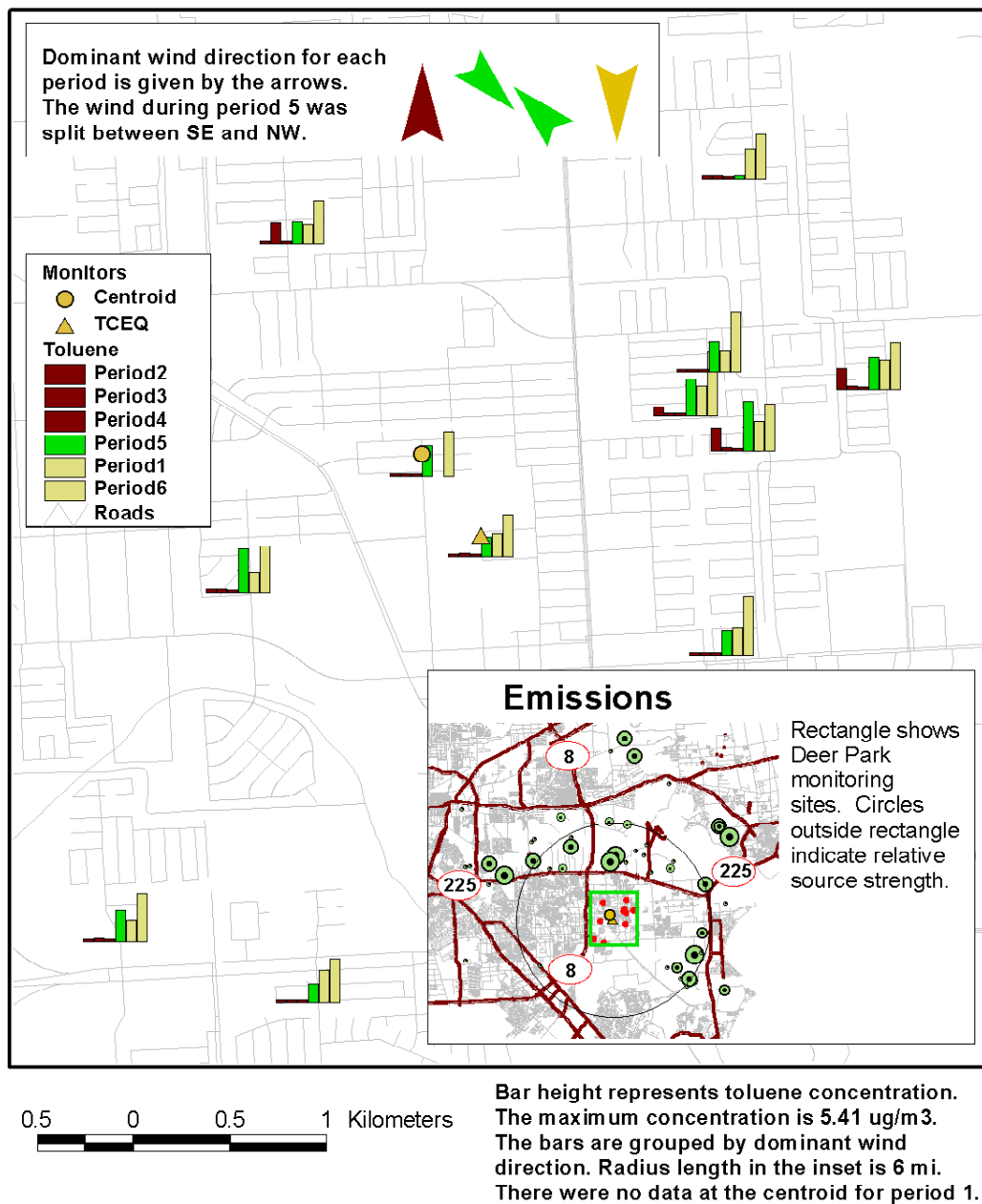


Figure 15. Toluene concentrations and emission sources in the Deer Park area of Houston, Texas.

m,p-Xylene Concentrations in Deer Park

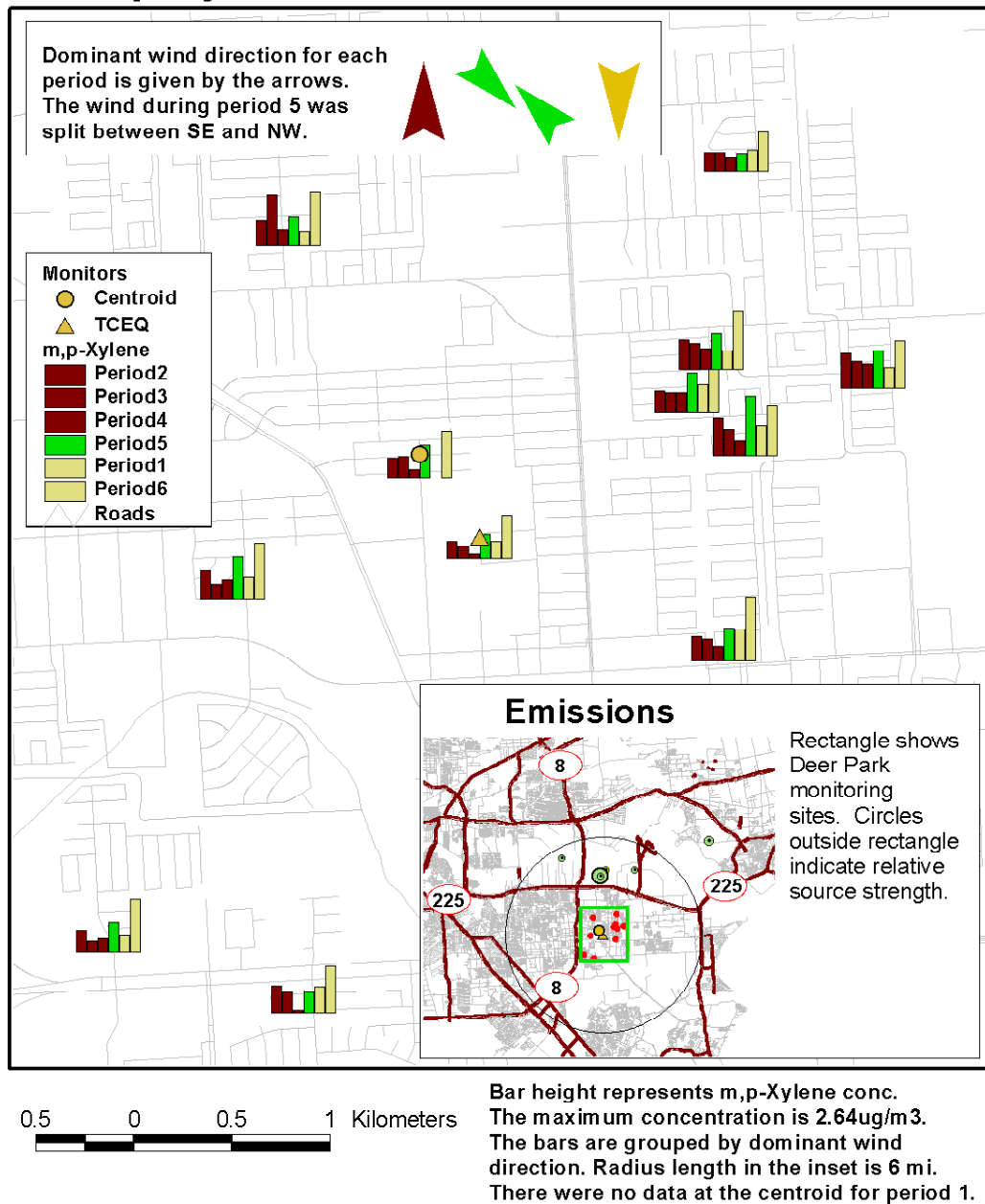


Figure 16. m,p-xylene concentrations and emission sources in the Deer Park area of Houston, Texas.

Chloroform Concentrations in Deer Park

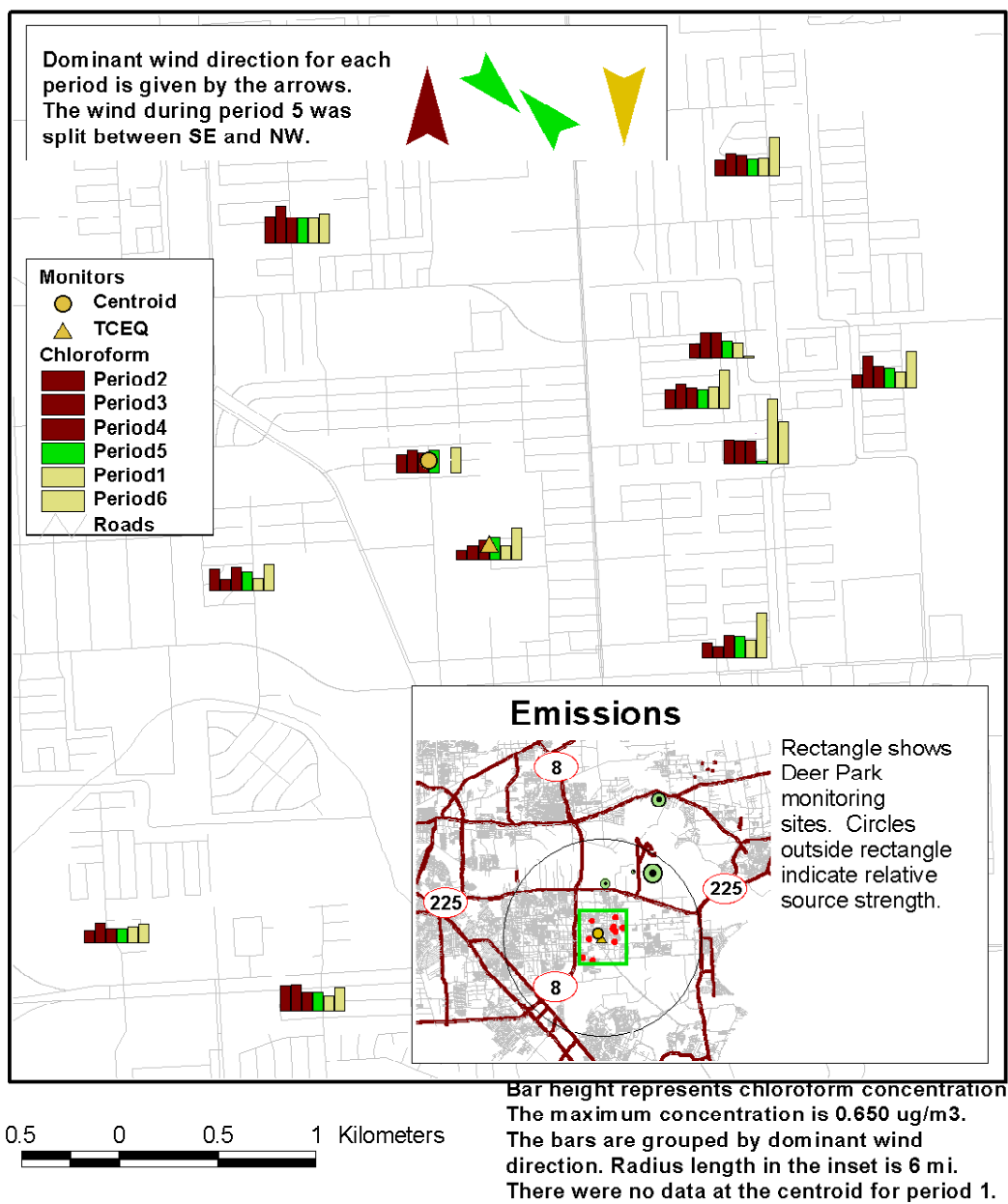


Figure 17. Chloroform concentrations and emission sources in the Deer Park area of Houston, Texas.

Carbon Tetrachloride Concentrations in Deer Park

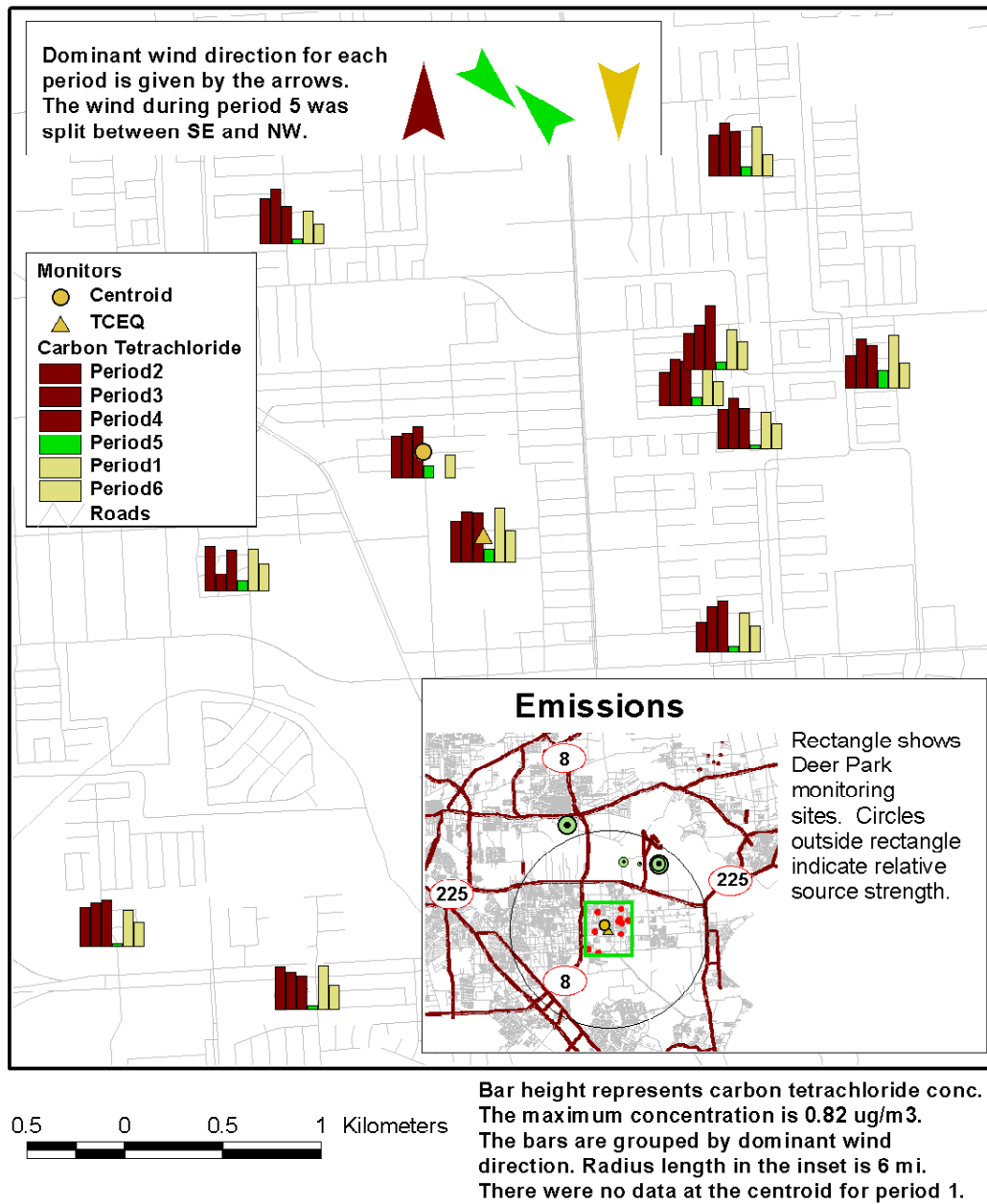


Figure 18. Carbon tetrachloride concentrations and emission sources in the Deer Park area of Houston, Texas.

n-Hexane Concentrations in Deer Park

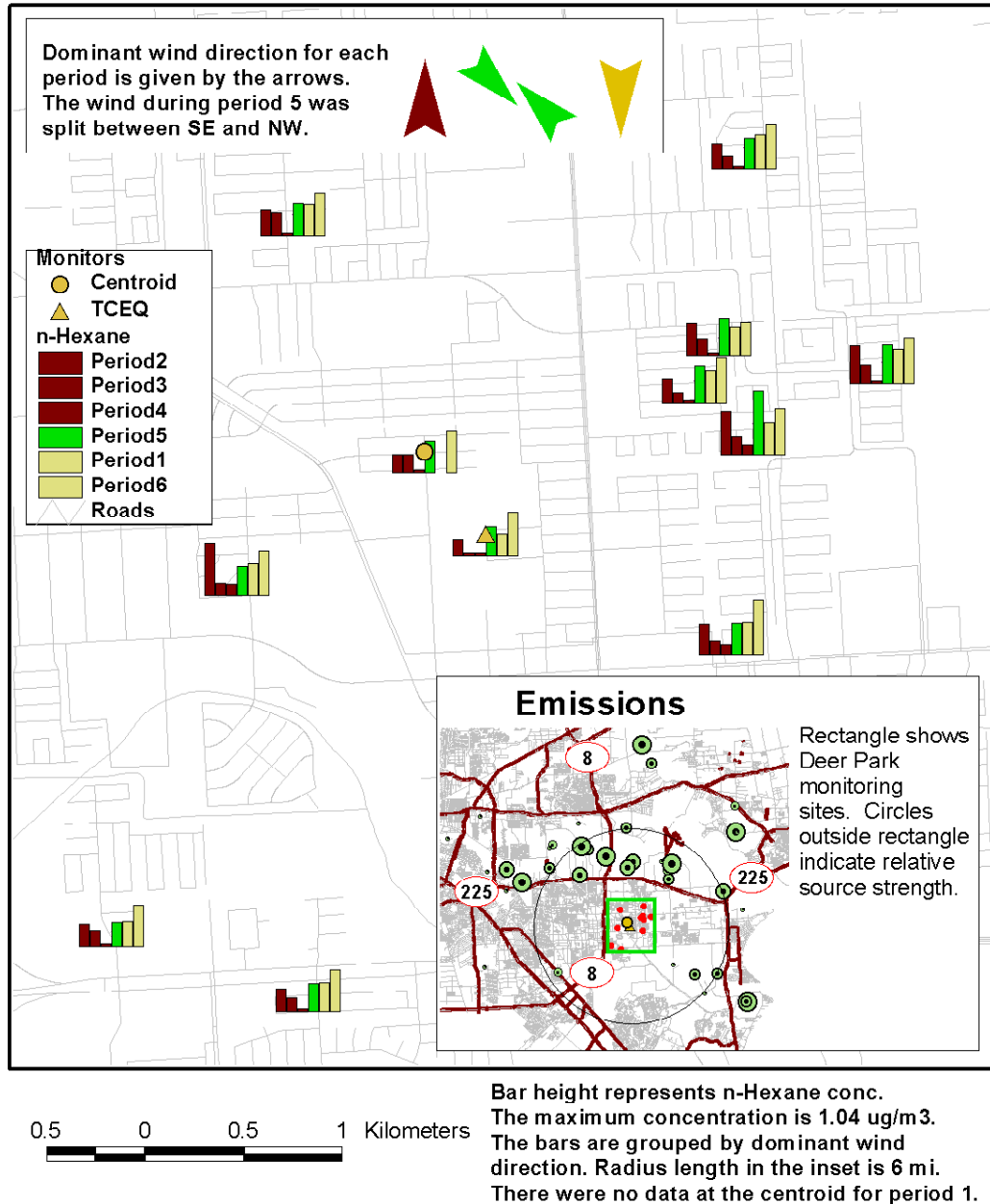


Figure 19. n-hexane concentrations and emission sources in the Deer Park area of Houston, Texas.

1,2,4-Trimethylbenzene Concentrations in Deer Park

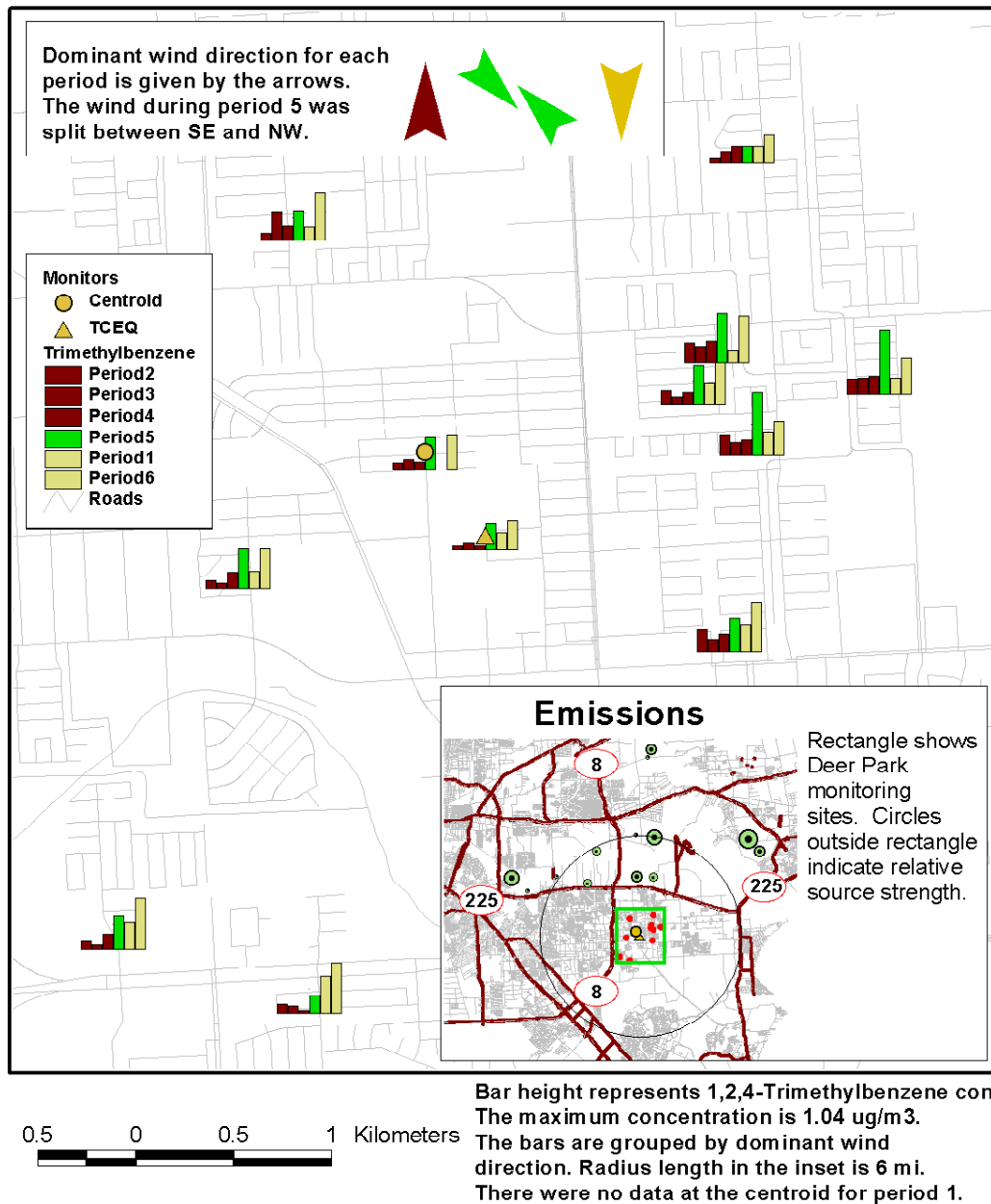


Figure 20. 1,2,4-trimethylbenzene concentrations and emission sources in the Deer Park area of Houston, Texas.

MTBE Concentrations in Deer Park

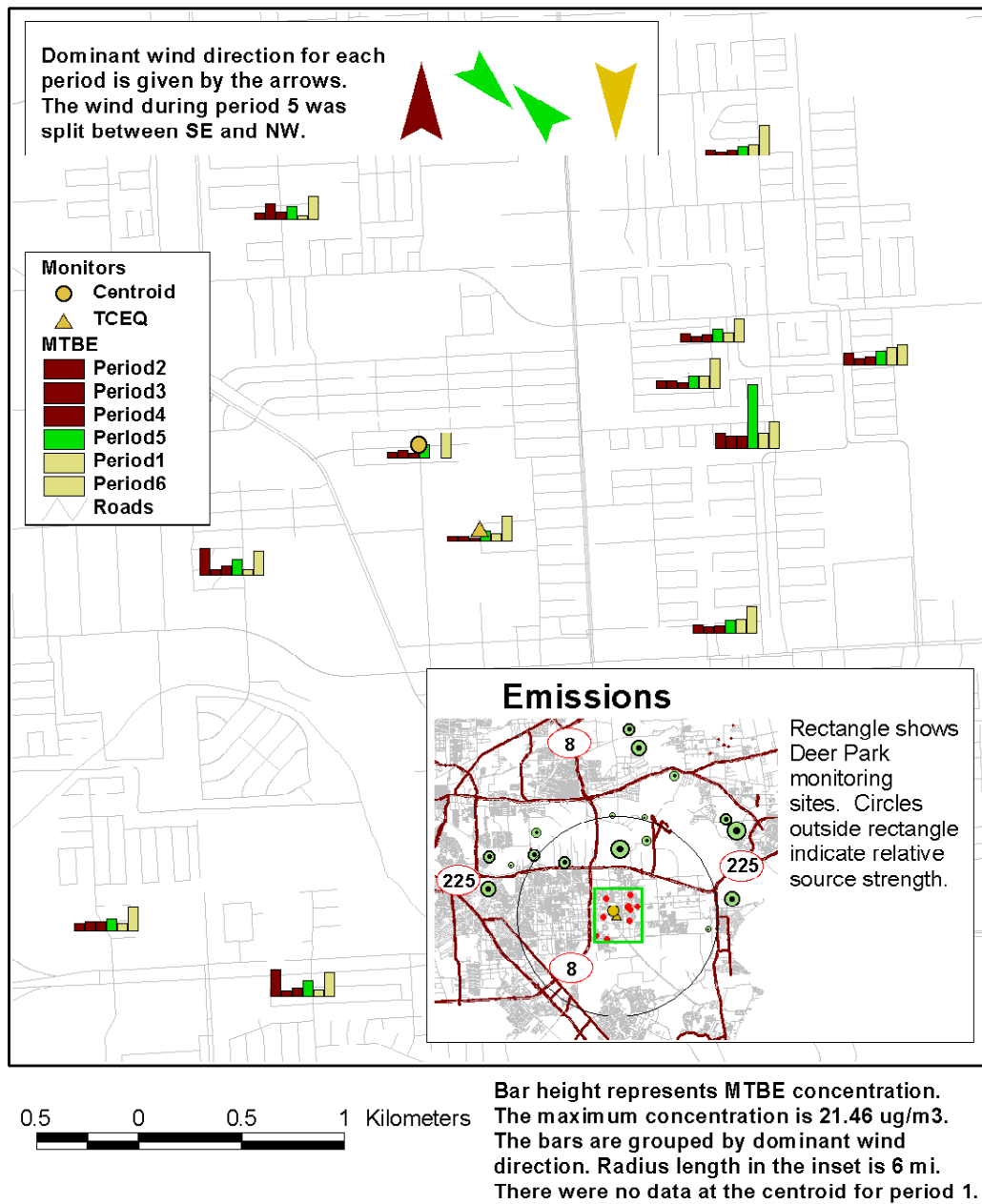


Figure 21. MTBE concentrations and emission sources in the Deer Park area of Houston, Texas.